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INVESTIGATION OF GAS TEMPERATURE MEASUREMENTS USING ULTRAVIOLET EXCITATION

A FINAL TECHNICAL REPORT

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ABSTRACT

A high resolution spectrographic study has been made of the nitrogen first negative system as induced by ultraviolet radiation. The radiation used in this experiment was the neutral helium resonance radiation at 584 Å. Nitrogen molecules at approximately 300°K and at a pressure of one Torr were ionized by the ultraviolet radiation with the aid of a windowless experimental configuration. The resulting nitrogen fluorescence was analyzed with sufficient resolution to enable study of the intensities of the R branch rotational lines of the 0-0 vibrational band of the first negative system. The data indicate that the induced population of the upper levels approximates a boltzmann distribution at the ambient temperature of the gas.

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INTRODUCTION

Temperature measurements using the high energy electron beam probe technique have certain advantages when studying low density gaseous flows. One important aspect is that no physical probe is introduced into the flow and therefore the medium is "undisturbed", allowing a more nearly exact temperature determination.

The validity of the method relies upon direct excitation of the gas molecules by the primary electron beam electrons for which "optical selection rules" apply, and that the lower energy secondary electrons (those electrons removed from the gas molecules in the direct beam excitation process) do not contribute to additional excitation of the gas molecules.

A controlled experiment using ultraviolet photons rather than the high energy electron beam should be able to provide some information concerning the excitation processes of the gas molecules. The advantages of such a study are two-fold. "Optical selection rules" obviously are valid for the electromagnetic radiation in the ultraviolet region and the energy of the "secondary" electrons (the ionized electrons) is known through conservation of energy considerations. The wavelength (and therefore energy) of the ultraviolet radiation can be chosen such that the "secondary" electron has insufficient energy to contribute in further gas molecule ionization and thus only the direct ionization process can contribute to the data leading to a temperature determination for the gas. Since the neutral

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helium resonance radiation at 584 \AA has the desired energy, and the (0, 0) band of the first negative system of N_2^+ has been used by a number of investigators for temperature measurements, a controlled laboratory investigation of the photoionization of the nitrogen molecule by that helium resonance radiation was initiated.

Previous laboratory studies of this process have been conducted under low spectroscopic resolution in which narrow bandpass filters were used to analyze the resulting molecular nitrogen ion fluorescence. These attempts were sufficiently adequate to obtain the gross features of the vibrational spectra of the nitrogen first negative system. Other techniques which relied upon measuring the energies of the secondary electrons of the photoionization process to determine the relative populations of the molecular ion electronic states have also been made, but these studies do not provide resolution of the rotational structure involved in the energy transfer process.

This investigation was conducted in an effort to study the N_2 fluorescence as produced by the incident He resonance radiation and to resolve the rotational lines of the 0-0 vibrational band of the first negative system. The fluorescence data obtained was compared with that produced by a Boltzmann distribution within the upper $v = 0$ vibrational level of the first negative system.

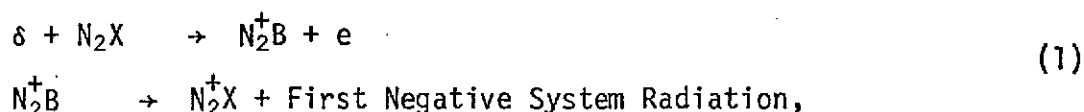
THEORY

To investigate the intensity distribution of the first negative system we note that incident ultraviolet radiation on N_2 may ionize the molecule and leave it in the ionic ground or excited state if the energy of the photon is greater or equal to the energy difference between the ionic state and the initial molecular state. Should the molecule be ionized then the emitted photoelectron carries off any excess energy. In the case of incident He resonance radiation at 584 \AA , the photon energy is sufficient to excite the $\text{N}_2^+ B$ state of the ion and the photoelectron is left with insufficient energy to ionize other nitrogen molecules that would contribute to the production of the first negative system. Therefore, we consider the specific

case for production of nitrogen's first negative system assuming a two step process:

- (a) Direct photoelectric excitation of N_2^+B from the molecular ground state N_2X and
- (b) The photoionized molecular ion produces the first negative system by spontaneous emission.

This ion subsequently recombines with the free electrons and cascades into the ground state of the neutral species. This may be represented by the following equations,



where δ is a photon of wavelength less than the N_2^+B excitation threshold of wavelength 691 Å.

The intensity of the fluorescence from the excited state N_2^+B is dependent upon the steady state population of the rotational levels of the various vibrational levels. The intensity of a particular rotational line is given by,

$$I = h\nu A_{JJ'} n_J \quad (2)$$

where n_J is the population of the upper rotational level and $A_{JJ'}$ is the transition probability for spontaneous emission from the rotational level J of N_2^+B to the level J' of N_2^+X . At sufficiently low temperature ($T \lesssim 300^\circ K$) only the $v = 0$ vibrational level of the neutral ground electronic state is significantly populated. Therefore, inspection of the Franck-Condon factors for $N_2X - N_2^+B$ shows that transitions only to the $v = 0$ vibrational level of N_2^+B will appreciably participate in the excitation process. Furthermore examining the Franck-Condon factors for the $N_2^+B - N_2^+$ transition shows that the maximum fluorescence will be observed for (0-0) vibrational transition.

In order to simplify the presentation of the calculations, we will not include the population alternation of the rotational levels caused by the presence of nuclear spin for the homonuclear molecule, and furthermore assume

a boltzmann distribution for the rotational levels of the ground electronic state. The population of the J^{th} rotational level of N_2X is then given by:

$$N_J = N_{\text{or}} \frac{(2J+1)}{(2J+1)} e^{-\beta J(J+1)} Q_r^{-1} \quad (3)$$

where N_{or} is the number of neutral N_2 molecules in the $v = 0$ vibrational level,

$$\beta = \frac{hcB_0}{KT}$$

and Q_r^{-1} is the partician function.

For a given vibrational level of $N_2^+B^2\Sigma_u$, J represents the total angular momentum. For the purposes of this treatment, we shall neglect the spin splitting of the rotational levels and assume the two spin levels degenerate and therefore sum over the two states in the calculations. This formally replaces J by K , the total angular momentum apart from the spin.

Then the population of the K^{th} excited level of N_2^+B is given by

$$N_K = N_{\text{ov}} \left[(K+1) e^{-\beta(K+1)(K+2)} + K e^{-\beta K(K-1)} \right] \quad (4)$$

which can be reduced to:

$$N_K = N_{\text{ov}} e^{-\beta K(K+1)} \left[(K+1) e^{-\beta(K+1)} + K e^{\beta K} \right] \quad (5)$$

Since for the specific case of $N_2 \times B_0 \approx 2.01 \text{ cm}^{-1}$ and assuming we can approximate N_K by

$$n_K = N_{\text{ov}} (2K+1) e^{-\beta K(K+1)}, \quad (6)$$

which should describe the population of excited states to within 2% as determined by comparison with Equation (4). It should be noticed that Equation (6) is a boltzmann distribution using $B = 2.01 \text{ cm}^{-1}$ of N_2X , rather than $B = 2.08 \text{ cm}^{-1}$ of N_2^+B in the evaluation. The intensity

distribution is therefore approximated by

$$I = h\nu A_{KK'} N_{ov} (2K + 1) e^{-BK(K + 1)} \quad (7)$$

where K' is the rotational quantum number of N_2^+X and $A_{KK'}$ is the spontaneous transition probability.

EXPERIMENTAL CONFIGURATION

The experimental configuration used for this experiment consisted of three basic elements: 1) the monochromatic ultraviolet radiation source, 2) the nitrogen gas test chamber, and 3) the fluorescence detection apparatus. Optical alignment of the entire configuration was obtained with the aid of a low power HeNe laser.

The monochromatic ultraviolet radiation source consisted of a modified McPherson Model 225 one-meter vacuum monochromator equipped with a Hinteregger discharge lamp mounted at the entrance slit. The monochromator was used with a MgF_2 overcoated concave grating blazed at 1500 \AA with a ruling of 600 l/mm. The exit slit assembly was specially fabricated to enable mounting of the test chamber with the slit area providing the only aperture between the two units. This configuration resulted in a relatively low pumping speed for gas flow between the test chamber and the main chamber of the monochromator.

The nitrogen gas test chamber was machined from aluminum with cylindrical geometry approximately 12.5 cm in diameter and a depth of 2 cm. The previously described exit slit of the one meter monochromator served as the entrance slit of this chamber and the opposite end of the chamber was equipped with a one-inch quartz viewing port. A nitrogen gas inlet port was also provided.

The fluorescence detection apparatus consisted of a one-meter JACO Czerny Turner type spectrograph Model 75-150 equipped with a grating blazed at 1.3μ with a ruling of 1180 l/mm and a 2" x 10" plate camera.

The basic considerations of the operational procedure were: (a) To maintain high enough pressure in the lamp; to produce sufficient intensity

of 584 Å, (b) low pressure in the path within the monochromator, the reduce absorption and prevent diffusion pump back-streaming, and (c) to keep sufficient pressure in the nitrogen chamber to maximize the intensity of the fluorescence.

The configuration adopted enabled windowless operation of a Hinteregger discharge lamp at approximately one Torr He pressure. A pressure differential was maintained between the lamp and the main chamber by restricting the He flow into the monochromator to an opening defined by the monochromator's entrance slit which was 150 μ wide and 5 mm high. These operating conditions together with continuous pumping by a six-inch diffusion pump, resulted in maintaining the pressure in the main chamber below 5×10^{-5} Torr. The nitrogen test chamber with a slit 100 $\mu \times 10$ mm was maintained at one Torr pressure for optimum operating conditions. This nitrogen pressure, together with the operating pressure in the Hinteregger lamp, resulted in a main chamber pressure of approximately 10^{-4} Torr. Tests indicated that at this pressure the ultraviolet radiation entering the test chamber was reduced by only a few percent over that of a relatively hard vacuum of less than 10^{-6} Torr.

The Hinteregger Lamp was operated in the DC mode at approximately 1700 V and 200 ma. The one-meter vacuum monochromator was set at 584 Å in first order. The fluorescence observing spectrograph was set in third order for recording the 0-0 band of the first negative system of nitrogen. This gave a linear dispersion of approximately 5 Å/mm, which was sufficient to resolve the rotational lines with entrance slit widths of 150 μ . Because the total fluorescence intensity of the 0-0 band was exceedingly weak in this experiment, spectroscopic 103 a0 plates were chosen for detection because of their relative resistance to reciprocity failure. These plates were exposed up to 18 hours for maximum density. The full slit height of the spectrograph was utilized in this experiment in order that a four-step neutral density filter could be in place during the actual exposure so that calibration could be included with the actual experimental data. Uniformity along the vertical length of the slit was ascertained through prior calibration.

CONCLUSIONS

Inspection of the spectrographic plate of the 0-0 vibrational band of

the first negative system of nitrogen as excited by the 584 Å He resonance radiation shows that the rotational lines of the R branch are clearly resolved, while the P branch forms the band head. The He 3888 Å line is used as a reference. A microdensitometer trace of the plate was made with the help of a Joyce Lobel instrument. The structure of the R branch is clearly seen with the 2:1 intensity alteration of the lines as well as the peak line intensities corresponding to that which would be expected at approximately 300°K temperature.

The 0-1 vibrational band has also been observed but of much less intensity, in agreement with the Franck-Condon principle for the first negative system.

The comparison of the observed intensity distribution with that prescribed from equation (4) shows reasonable agreement, in that both the peak intensities appear at the 6-7 transition and that fitting the points to a boltzmann distribution provides a temperature measurement of $320 \pm 20^\circ\text{K}$. Further investigation should be made at lower temperature to confirm the rotational distribution produced by the ultraviolet excitation process. A lower temperature should shift the intensity peak toward lower rotation lines of the first negative system and increase the intensity of the stronger lines thereby reducing the margin of error.